ATTACHMENT II - MELPIGNANO et al., Journal Article

Efficient light extraction and beam shaping from flexible, optically integrated organic light-emitting diodes

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Efficient out-coupling light extraction and optical beam shaping have been combined to form an integrated, flexible illuminator that is based on organic light-emitting diodes (OLEDs). Spherical refractive microlenses were replicated by a cost-effective UV-casting technique onto a plastic foil to achieve the customized Gaussian distribution of the electroluminescence of an OLED pixel (50 μm) matrix. The fabricated optical device, namely a ceiling light illuminator, shows an improvement of 70% in the out-coupled emission measured in the far field. The result corresponds to the theoretical prediction for isotropic emitting sources. © *2006 American Institute of Physics.*
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Since the first observation of efficient electroluminescence (EL) emission from an organic thin-film double layer heterostructure[1] and from a single layer conjugated polymer,[2] organic light-emitting diodes (OLEDs) have attracted a great deal of attention as promising candidates for low-cost, highly innovative, flat-panel displays. This promise has already been fulfilled and small size displays using OLEDs as the active element are actually on the market. Recently, OLEDs have achieved a current density of several hundred ampere per square centimeter.[3] This impressive result opens the way to the possibility of electrically pumped organic laser diodes. Another approach to this goal is being pursued by combining charge transport and light emission in ambipolar organic materials into organic light-emitting transistor architectures (OLETs).[4–8] However, there are still unsolved problems, both fundamental and technological, in the full exploitation of the unique properties of organic systems as light sources. OLEDs can be viewed as energy conversion devices (electricity to light) based on EL and it is well documented in the literature how "state of the art" OLED devices, namely based on electrophosphorescent organic materials and engineered charge-carrier balance, are able to approach 100% internal quantum efficiency.[9,10]

Despite those remarkable achievements, OLED external light efficiency is only a few percent due to unresolved problems in the efficient and cost-effective light extraction from OLED devices. This is a critical point which has to be addressed and resolved, in particular for the realistic exploitation of OLEDs as innovative solid-state light sources for general lighting applications and related technologies.[11] In this perspective, OLEDs are promising candidates to effectively compete with incandescent, fluorescent, and light-emitting diodes sources. In this context we have addressed the problem of efficient light extraction in OLEDs in combination with the control of the far-field light distribution. A flexible, optically integrated OLED illuminator is demonstrated with enhanced light extraction combined with a customized Gaussian far-field distribution of the EL emitted radiation, which conforms to high standard ceiling light illuminators for automotive applications.[12]

The main reason for the low out-coupling efficiency of standard OLEDs is losses due to total internal reflection and waveguiding effects.[13] This behavior has been modeled by simple ray tracing. Losses of up to 80% of the produced photons have been calculated.[14–16] In order to improve the out-coupling efficiency, different methods have been investigated: interfacial rough or textured surfaces,[17,18] reflecting surfaces and distributed Bragg reflectors,[19,20] as well as two-dimensional (2D) ordered photonic structures.[21,22] Recently, enhancement of the out-coupling OLED efficiency by creating an interface to a glass substrate with ordered microlenses arrays has been demonstrated to improve the out-coupling OLED efficiency by a factor of 1.5 (50%) (Ref. 23). From our point of view the optimization of the outward emission is only part of the problem since the far-field distribution and divergence, as well, need to be controlled for the car ceiling light. In particular, high intensity illumination in a given area with a specific intensity distribution is required. In order to satisfy the stringent automotive specifications for the far-field light distribution of such a device, the Lambertian emission profile from the OLED has to be transformed. This is achieved by collecting and redirecting the light from an array of small OLED pixels with matched microlenses arrays on the same substrate. In our case, the design constrains the optimized OLED pixel dimension to 50×50 μm (Ref. 24). The individual elements of the microlens array are slightly offset from their matched position in respect to the OLED pixel by a maximum amount of 15 μm. The far-field light distribution (see supplemental material) is obtained by the sum of overlapping images of the OLED pixels, which is called a patch pad design. The basic unit of 5×5 microlenses (see Fig. 1) forms the desired light distribution in the far field. This approach has two advantages: the emitted light

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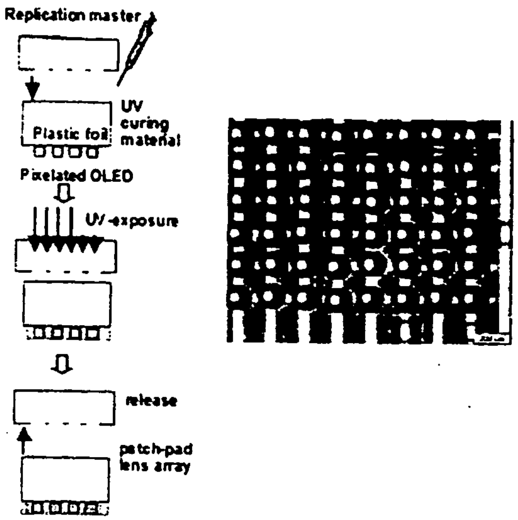

FIG. 1. (Color) Schematic of the UV-casting method for fabricating refractive micro-optics arrays on a flexible substrate. The picture shown on the right-hand side shows the obtained device. The ITO patterned stripes are clearly visible. The OLED pixel source is positioned underneath the refractive lens. The box highlights the engineered 5 × 5 optical unit producing the Gaussian light distribution.

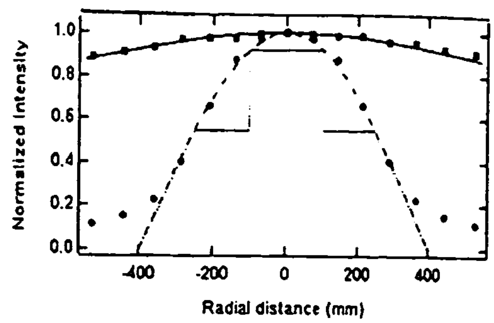

FIG. 2. Calculated and experimental optical field distribution generated by the engineered micro—optical pattern integrated to the OLED illuminator. The continuous curve corresponds to the ideal normalized Lambertian distribution. The squares represent the measured light distribution obtained by an OLED without microlenses; the dashed line is the best-fit Gaussian profile ($y=y_0+A\exp[-(x-x_0)/\sigma]2$, with $y_0=-1.617$, $A=2.614$, $x_0=6.355\,e^{-15}$, $\sigma=557.15$) of the ceiling light intensity distribution represented by the thin continuous line. The black dots represent the measured normalized intensity obtained by a micro-optics integrated OLED device. In both cases a very good correspondence between theoretical and experimental data is obtained.

is concentrated and shaped in a specific solid angle, and the light extraction from the OLED multilayer structure is improved by direct optical coupling.

The integration of refractive micro-optics by UV casting onto a suitable flexible substrate provided by Vitex[25] is sketched in Fig. 1. The desired pattern of microlenses was originated by photoresist reflow technology and then converted to a master mold in UV transparent silicon rubber (PDMS). Employing a UV curable material, the micro-optical structure is replicated onto the backside of a patterned indium-tin-oxide (ITO) Vitex flexible foil. A micrograph of the obtained optically integrated substrate is shown in the inset of Fig. 1. The OLED source was the classical small molecules heterojunction structure, vacuum grown in a simple architecture:[26] plastic substrate (125 μm), ITO (160 nm), α-NPB [N,N′-di (naphthalene-1-yl)-N,N′-diphenylbenzidine] (60 nm), Alq3 [tris-(8-hydroxyquinoline) aluminium] (70 nm), LiF (lithium fluoride) (0.8 nm), Al (aluminium) (100 nm).

The OLED pixel sources were mechanically aligned with a precision of 5 μm to the refractive micro-optics pattern. During the same deposition run a reference OLED sample, without a refractive micro-optics pattern integrated in the backside, was also fabricated for comparison.

In order to verify the agreement of the light distribution of the fabricated illuminators with the simulated one, the OLED samples with and without micro-optics have been measured by placing a photon counter in the far-field region of the optical pattern and performing an angular scan of the samples.

The experimental light distribution of the samples is compared with the theoretical one in Fig. 2.

A Lambertian profile is obtained for the device without micro-optics, as expected for a planar OLED, and very good agreement is obtained by our prototype ceiling light illuminator with the theoretical Gaussian optical distribution.

The second aim of our work is to significantly improve the light extraction efficiency keeping the architecture of the device as designed for optical field distribution control.

The OLED modeling for the evaluation of light out-coupling efficiency has been performed using the software ASAP and considering the spectral distribution of the emitted light. Some wavelengths of the emission spectrum are selected and inserted with their relative weights into ASAP. For each medium (the investigated devices possess a two-layer structure), the refractive index, measured on thin-film samples by spectroscopic ellipsometry, corresponding to the previously entered wavelength, was inserted. The polychromatic emission was then simulated by automatic interpolation of the weights of the wavelengths between the selected ones and by interpolation of the measured refractive indices. Depending on the isotropic or Lambertian pattern used to simulate the emitting source, a remarkable quantitative difference of the extraction out-coupling efficiency has been found. The predicted results are reported in Table I.

The simulated results were compared with the extraction efficiency of the real devices. Both the illuminators with and without integrated micro-optics were placed into an integrating sphere for *L-I-V* characterization and quantitative measurements. The spectral distribution and consequently the color rendering of the two samples were identical. The quantitative results obtained from *L-I-V* curves are reported in Fig. 3. The experimental results are in good agreement with the model and show an improved extraction efficiency of 70% for the integrated micro-optics controlling the light distribution as a Gaussian pattern. The experimental results in-

TABLE I. Theoretical improvement in light extraction with an isotropic and a Lambertian emitter.

Frontal light extraction		
	Isotropic emitter	Lambertian emitter
Flat air-substrate interface	15.15%	27.55%
Microlenses air-substrate interface	25.77%	41.70%
Δ% light extraction	70%	51.3%

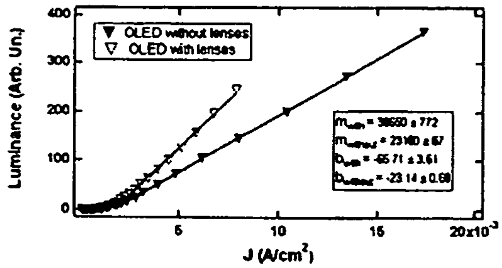

FIG. 3. Luminance vs current density for the OLED illuminator with and without integrated refractive micro-optics. The modeled light extraction improvement can be evaluated by the ratio of the angular coefficient (m) of these fitted lines. In our case the ratio $\Gamma = m_{with} - m_{without} / m_{without} = 67 \pm 1\%$ obtained by the experimental curves is in good agreement with the theoretical prediction of 70% for the isotropic source model.

dicate the isotropic nature of the emitting OLED source (see Table I), which is consistent with the theory of radiative excitonic decay generated by electron-hole recombination into the emitting Alq3 moiety. The external Lambertian distribution measured on planar OLEDs originates from the interaction of isotropically emitted radiation from excitonic emitting species with the multilayered device architecture. In conclusion, we have demonstrated how a simple and competitive technique, based on UV casting, can integrate suitable refractive micro-optical lens arrays onto a flexible substrate to control the desired light distribution, in our case a Gaussian beam shape, emitted by a vacuum sublimed OLED matrix. Together with the optical field distribution control, a 70% gain in outward light extraction efficiency opens the way to possible full exploitation of OLED light sources as efficient, energy- saving illuminators for general lighting and for automotive applications.

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[1]S. W. Tang and S. A. Van Slyke, Appl. Phys. Lett. 51, 913 (1987).
[2]J. H. Burroughes, D. D. C. Bradley, A. R. Brown, R. N. Marks, K. Mackay, R. H. Friend, P. L. Burns, and A. B. Holmes, Nature (London) 347, 539 (1990).
[3]H. Nakanotani, H. Sasabe, and C. Adachi, Appl. Phys. Lett. 86, 213506 (2005).
[4]A. Hepp, H. Heil, W. Weise, M. Ahles, R. Schmechel, and H. von Seggern, Phys. Rev. Lett. 91, 157406 (2003).
[5]C. Rost, S. F. Karg, W. Riess, M. A. Loi, M. Murgia, and M. Muccini, Appl. Phys. Lett. 85, 1613 (2004).
[6]F. Cicoira, C. Santato, F. Dinelli, M. Murgia, M. A. Loi, F. Biscarini, R. Zamboni, P. Heremans, and M. Muccini, Adv. Funct. Mater. 15, 375 (2005).
[7]C. Santato, I. Manunza, A. Bonfiglio, F. Cicoira, P. Cosseddu, R. Zamboni, and M. Muccini, Appl. Phys. Lett. 86, 141106 (2005).
[8]M. A. Loi, C. Rost-Bietsch, M. Murgia, S. F. Karg, W. Riess, and M. Muccini, Adv. Funct. Mater. 16, 41 (2006).
[9]M. A. Baldo, D. F. O'Brien, Y. You, A. Shoustikov, S. Sibley, M. E. Thompson, and S. R. Forrest, Nature (London) 395, 151 (1998).
[10]C. Adachi, M. A. Baldo, M. E. Thompson, and S. R. Forrest, J. Appl. Phys. 90, 5048 (2001).
[11]In a recent report of the U.S. Department of Energy it is pointed out how in the U.S. 20%, and worldwide 10%–15%, of the total production of electricity is consumed for operation of lighting systems (OIDA Report, August 2002). It appears clear how for a sustainable economy and for environmental aspects a worldwide priority is to reduce the energy consumed for general illumination. It is also clear how the major enabler of energy conservation will be a new lighting technology such as solid-state lighting.
[12]P. Melpignano, S. Sinesi, G. Rotaris, M. Antonipieri, F. Cicoira, M. A. Loi, M. Muccini, R. Zamboni, M. T. Gale, and S. Westenhoefer, Synth. Met. 139, 913 (2003).
[13]V. Bulović, V. B. Khalfin, G. Gu, P. E. Burrows, D. Z. Garbuzov, and S. R. Forrest, Phys. Rev. B 58, 3730 (1998).
[14]N. C. Greenham, R. H. Friend, and D. D. C. Bradley, Adv. Mater. (Weinheim, Ger.) 6, 491 (1994).
[15]G. Gu, D. Z. Garbuzov, P. E. Burrows, S. Venkatesh, S. R. Forrest, and M. E. Thompson, Opt. Lett. 22, 396 (1997).
[16]N. Tessler, Appl. Phys. Lett. 77, 1897 (2000).
[17]I. Schnitzer, E. Yablonovitch, C. Caneau, T. J. Gmitter, and A. Scherer, Appl. Phys. Lett. 63, 2174 (1993).
[18]B. J. Matterson, J. M. Lupton, A. F. Safonov, M. G. Salt, W. L. Barnes, and I. D. W. Samuel, Adv. Mater. (Weinheim, Ger.) 13, 123 (2001).
[19]I. Schnitzer, E. Yablonovitch, C. Caneau, and T. J. Gmitter, Appl. Phys. Lett. 62, 131 (1993).
[20]F. A. Kish, Jr. and S. A. Stockman, US Patent No. 6 015719 (2000).
[21]Shanhui Fan, Pierre R. Villeneuve, J. D. Joannopoulos, and E. F. Schubert, Phys. Rev. Lett. 78, 3294 (1997).
[22]M. Borodistky, R. Vrijen, T. F. Krauss, R. Coccioli, R. Bath, and E. Yablonovitch, J. Lightwave Technol. 17, 2096 (1999).
[23]S. Moeller and S. R. Forrest, J. Appl. Phys. 91, 3324 (2002).
[24]It is worth noting how, in spite of the incoherent nature of the EL, a partial coherence can be obtained by spatial constrain of the emitting OLED pixel. In our case we have found that in the plane of micro-optics the maximum radius of spatial coherence is about 1.5 μm. However, changing the pixel dimension and the substrate thickness it is possible to increase this number. This is interesting since it may be exploited for diffractive optics integration in OLED devices opening the way to an alternative optical integration technology.
[25]Vitex Systems, 3047 Orchard Parkway, 95134 San Jose, CA, USA.
[26]C. W. Tang, S. A. Van Slyke, and C. H. Chen, Appl. Phys. Lett. 65, 3610 (1989).